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Diagenesis of the Skeletal Remains in Four Archaeological Sites in Northern Jordan

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Abstract

Archaeological bone chemistry studies from Jordanian sites have overlooked the important role of bone diagenesis, which may rule out proper interpretation of data. This study evaluates bone diagenesis in 81 human and animal bone samples from four Byzantine sites in northern Jordan. The evaluation of diagenesis was structured on the deviations of bone chemistry from a set of standards using XRD, XRF, and AAS. The results show significant diagenesis in the four sites, which consequently biases reconstructions of paleodiet using trace element analysis. The study also shows that burial type and environment are major factors in the process of diagenesis.

Keywords: Skeletal Remains, Archaeological Sites.

Introduction

Trace elements analysis of archaeological human bones has been used since the late 1980s to reconstruct past human diet (Buikestra et al. 1989). Most of those studies use Sr/Ca and Zn/Ca ratios in the bone and/or teeth apatite as indicators of plant and animal food consumption (Sponheimer et al. 2005). The results would be persuasive if post mortem contamination of bones is evaluated and the extracted data calibrated before any interpretation is made (Edward and Benfer 1993; Zapata et al. 2006). Postmortem contamination or diagenesis is "the cumulative physical, chemical, and biological processes that alter all archaeological materials in the burial environment; these processes will modify an inorganic object's original chemical and/or structural properties and will govern its ultimate fate, in terms of preservation or destruction" (Wilson and Pollard 2002). Diagenesis is multi-

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factorial, where the local geology (Edward and Benfer 1993), land use (Grupe et al. 1997; Jans et al. 2002), burial environment (Ezzo 1997; Grupe et al. 1997) and mineralogical composition of the surrounding soil (Stephan 1997) may alter the mineralogical composition of buried bones. Bioarchaeological studies in the region have ruled out the role of diagenesis and soil/bone exchange in optimizing the accuracy of the extracted data (Grattan et al. 2002; Payatt et al. 2000; Perry et al. 2008; Shdouh 2003), which calls into question the interpretations and the overall archaeological context in those studies. Furthermore, the trace elements in archaeological bones might be biogenic in nature if present in certain concentrations otherwise of a diagenetic origin (Martinez-Gracia et al. 2005; Millard 2006).

Archaeological bone is composed of an organic material called collagen and an inorganic material called hydroxyapatite [$Ca_{10}(PO_4)_6(OH)_2$], where most of the paleodietary reconstructions take place. Table 1 below shows some of the possible substitutions at the Ca, PO_4 and OH sites (Legeros et al. 1980; Young 1975) that alter the chemical composition of buried bones.

Table 1: The major substitutions in the hydroxyapatite structure

Ca site	PO_4 site	OH site
Sr^{2+}	CO_3F^{3-}	CO_3^{2-}
Pb^{2+}	AsO_4^{3-}	S_2^{2-}
Mg^{2+}	VO_4^{3-}	O^{2-}
Ba^{2+}	CO_3F^{3-}	F^-
Na^{2+}	SiO_4^{4-}	Cl^-
	HPO_4^{2-}	O_2^-
	CO_3^{2-}	H_2O
		N_2

A bone sample that has post mortem contamination may have a Ca/P ratio of more than 2.15, a Ca content of more than 38%, a P content of less than 18%, an Sr concentration of more than 1000 ppm, a Pb concentration of less than 100 ppm, an Al concentration of less than 20 ppm, a Zn concentration of less than 200ppm,

or an Mn concentration of less than 10 ppm (Table 2). It has been shown that shallow burials and burials close to groundwater may add post mortem contamination (Bell, Skinner and Jones 1996).

Table 2: The reference values of trace elements in modern human bones (uncontaminated)

Minearology	Value	Reference
Ca/P	≈2.15	(Sillen 1981); (White & Hannus 1983)
Ca	≈38%	(Zapata et al. 2006)
P	≈18%	(Zapata et al. 2006)
Sr	<1000ppm	(Kyle 1986;Radosevich 1993)
Pb	≤ 100ppm	(Zapata, Pereze-Sirvent, Martinez-Sanchez, & Tavor 2006)
Al	≤20ppm	(Zapata et al. 2006)
Zn	<200ppm	(Zapata et al. 2006)
Mn	≤10ppm	(Zapata et al. 2006)

Sa‘ad, Ya‘amun, Juhfyiah, and Bersenia are the main archaeological sites in the north of Jordan that yielded enormous amounts of human and animal bones. The recovered faunal materials were dated to the Late Roman/Early Byzantine period except for Juhfyiah, which was dated to the Persian period. These sites have the potential to shed light on the paleodiet of the Byzantine period and thus allow more inferences about the socioeconomic status of the inhabitants and in a broader sense the role of settlement. The current study, thus, provides a method for investigating the skeletal remains chosen for reconstructing paleodiet. It hypothesizes that the skeletal preservation among those sites is as varied as the geological settings, and that the type of burial plays a major role in the preservation of the skeletal remains.

Materials and methods

The bone samples belong to four cemeteries at the archaeological sites of Ya‘amun, Sa‘ad, Bersenia and Juhfyiah in northern Jordan (Fig. 1). All of the

collected samples belong to adult humans except for the site of Juhfyiah, where animal bones were collected as a control sample because the diet of mammals is assumed to be known (Gonzales-Reimers et al. 2003; Price 1985) and thus diagenetic effects could be evaluated clearly.

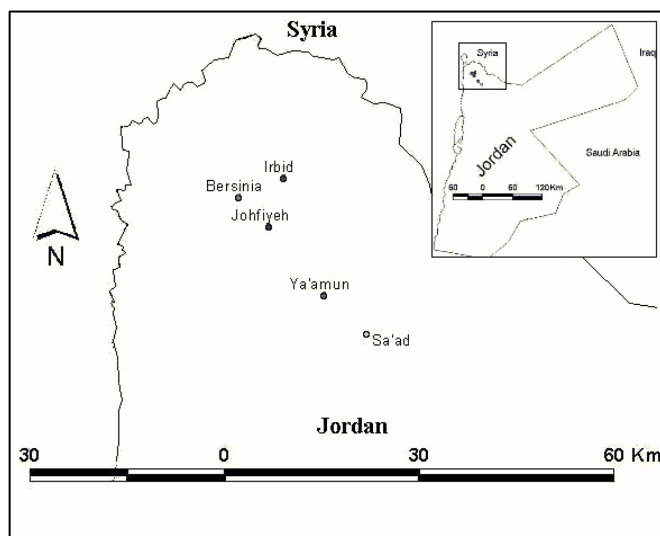


Fig. 1: The location of the sampled archaeological sites

The samples included a total of 70 human bones and 11 animal bones (Ovis/Capra) as well as a burial soil sample from each site. The samples were taken from the diaphysis of the mature long bones because they are the least susceptible to diagenesis (Ezzo 1994; Grupe and Piepenbrink 1988) and represent decades of the average intake of trace elements (Baraybar and de la Rua 1995). Immature long bones have a thinner cortex and lesser mineralization, which consequently may add bias as they are inherently more susceptible to diagenesis (Edward and Benfer 1993). None of the sampled bones had been treated with consolidants or preservatives.

The trabecular bone was completely removed to eliminate any trapped soil particles. The outer and inner surfaces of the remaining compact bone were mechanically cleaned with a diamond bit to remove any adhering soil. The samples were then cleaned with distilled water and left to dry at room temperature for 24 hours. All of the samples were ground to a fine powder using an agate mortar and a pestle. Atomic Absorption Spectroscopy (UNICAM 939) was used to

analyze the elements of Ca, Sr, Mn, Pb and Al in the bone samples, while the Ca/P ratio was determined using X-ray fluorescence (MiniPal2 Pananalytical). The mineralogical composition of samples (bones and soil) was determined using X-ray diffraction (Shimadzu 6000).

Table 3: Burial types and sample total numbers of the archaeological sites

Site	Samples no. Trace elements	Samples no. XRF	Samples no. XRD	Burial type
Ya'amun	20	7	7	Cave tomb filled with soil
Sa'ad	20	11	11	Horizontal shaft tomb
Bersenia	30	8	8	Horizontal shaft tomb filled with soil
Juhfyiah	11	6	6	Soil pit
Total	81	32	32	

Results and discussion

The samples from the four sites yielded Ca/P ratios between 2.66 and 3.11, significantly above the referenced value of 2.15 as shown in Table 4 below. That means that the skeletal materials from all of the sites were subjected to diagenesis. The bones from Tell Juhfyiah demonstrate the greatest diagenesis as they probably were buried in soil pits, which were susceptible to water seepage from the surface runoff, enhancing the entrance of calcium into the bones or from ground water as the only site that revealed Fluoroapatite in bones. The least diagenesis is in the Sa'ad bones; the horizontal shaft tombs in Sa'ad had low amounts of soil fill and thus minimum soil/bone exchange. The values of Ca/P in both Ya'amun and Bersenia support our argument that the filled horizontal shaft tombs in Bersenia had Ca/P values greater than the unfilled ones in Sa'ad.

Table 4: Calcium and Phosphorus concentrations in bones in (%) using X-ray fluorescence

Sample no.	Sa‘ad		Ya‘amun		Juhfyiah		Bersenia	
	Ca	**P**	**Ca**	**P**	**Ca**	**P**	**Ca**	**P**
1	67.80	27.00	72.00	24.00	72.00	23.00	70.00	25.00
2	69.30	25.00	71.70	23.00	71.20	24.00	69.60	26.00
3	69.60	25.00	70.80	25.00	74.20	20.00	69.20	26.00
4	69.60	26.00	70.60	25.00	73.50	22.00	70.00	29.00
5	70.20	26.00	70.90	24.00	71.40	24.00	69.40	27.00
6	68.70	27.00	71.80	23.00	70.00	26.00	70.10	25.00
7	69.70	26.00	70.50	25.00			71.00	24.00
8	69.20	26.00					70.00	24.10
9	68.90	26.00						
10	69.60	26.00						
11	69.50	26.00						
Average	69.28	26.00	71.19	24.14	72.05	23.17	69.91	25.76
Ca/P	2.66		2.95		3.11		2.71	

The X-ray diffraction of the samples shows that they gained Ca mainly from gypsum, calcite and faujasite in proportions ranging from 4.32 to 14.93% for gypsum, 5.72% for calcite and 17.44% for faujasite. There are other minerals that were incorporated into the bone samples such as kaolinite, fluoroapatite, cordierite and sepiolite. Tell Juhfyiah is an artificial mound created thousands of years ago by cumulative human activities and most of its soil is actually ash, which explains the absence of gypsum, calcite and sepiolite in the buried bones.

The higher amounts of gypsum in the bones from Bersenia and Ya‘amun are due to the presence of wet soil around the bones as explained previously. Jordanian soil in general is very rich in $CaCO_3$, which reacts with sulphate (produced by sulphate-producing bacteria) in the presence of water yielding gypsum $(CaSO_4.2H_2O)$ according to formula 1 and 2 below:


$SO_3 + H_2O \rightarrow H_2SO_4$ …. (1)


$CaCO_3 + H_2SO_4 \rightarrow CaSO_4.2H_2O + CO_2$ …. (2)

Table 5: Mineralogical composition of bone in the four sites (in %), calculated from the XRD peaks

Site	Hydroxyapatite $Ca_{10}(PO_4)_6(OH)_2$	Kaolinite $Al_2Si_2O_5(OH)_4$·	Faujasite $Na_2,Ca,Mg)_3.5[Al_7Si_{17}O_{48}]·32(H_2O)$	Fluoroapatite $Ca_{10}(PO_4)_6F_2$	Gypsum $CaSO_4·2H_2O$	Calcite $CaCO_3$	Cordierite $Mg_2Al_4Si_5O_{18}$	Sepiolite $Mg_4Si_6O_{15}(OH)_2 • 6H_2O$	%
Juhfyiah	41.44	21.57	17.45	19.54	-	-	-	-	100
Bersenia	58.13	23.64	-	-	18.23	-	-	-	100
Sa‘ad	56.91	33.05	-	-	4.32	5.72	-	-	100
Ya‘amun	46.77	14.43	-	-	14.91	-	14.93	8.96	99.96

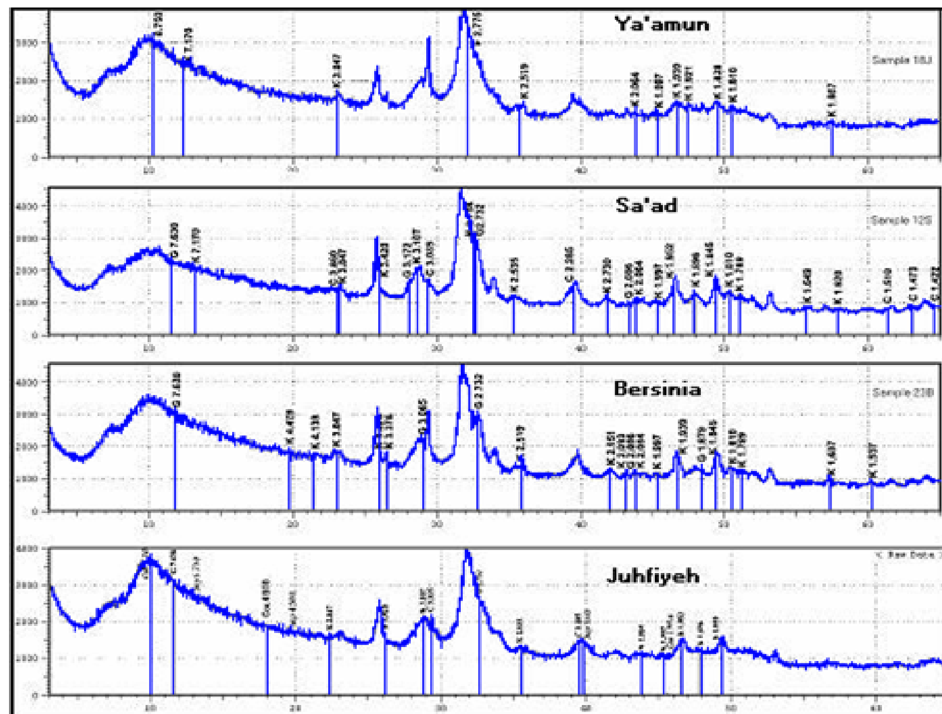


Fig. 2: X-ray diffraction of the bone samples from the four archaeological sites. Unlabeled peaks are hydroxyapatite.

The average concentration of Sr in the four sites is very high compared to the values for modern bones (table 2), where the optimum Sr level usually does not exceed 1000 ppm even in vegetarians due to its physiological turn over. The site of Bersenia has a lead average of about 152.21 ppm, while it is absent in the other sites. Lead concentration in blood of more than 20 ppm is fatal (Edward and Benfer 1993) but may not reach a value above 100 ppm (Zapata, Pereze-Sirvent, Martinez-Sanchez and Tavor 2006); a person with lead poisoning is likely to die before lead concentration reaches the average value at Bersenia. Consequently the lead in the Bersenia bones was of a diagenetic nature. The elements of Zn, Mn and Al (Al is absent in the Bersenia samples) are very high in comparison to modern bone, signifying the prolonged exchange of those elements with the burial soil.

Table 6: Trace element results from Bersenia in ppm

Ca	Sr	Zn	Mn	Pb	Al
176900.00	534.40	130.20	59.99	12.48	0.00
337600.00	1373.00	193.90	99.28	61.97	0.00
320600.00	1043.00	177.10	31.58	138.10	0.00
258300.00	1206.00	226.30	77.07	134.40	0.00
374000.00	2124.00	205.70	83.21	13.32	0.00
361200.00	2695.00	292.60	43.80	40.16	0.00
348300.00	2044.00	215.20	252.60	70.64	0.00
320600.00	2479.00	244.40	296.60	162.90	0.00
257800.00	3130.00	230.30	235.20	221.70	0.00
357800.00	2846.00	179.70	145.10	35.50	0.00
334200.00	3067.00	196.40	234.00	179.80	0.00
365500.00	3747.00	212.00	294.10	139.50	0.00
300900.00	5915.00	152.60	416.30	396.20	0.00
287000.00	4181.00	186.70	264.30	471.10	0.00
302800.00	2318.00	337.60	214.10	150.40	0.00
425300.00	2963.00	211.50	220.10	155.40	0.00
289900.00	4167.00	118.20	306.00	332.70	0.00

Ca	Sr	Zn	Mn	Pb	Al
293400.00	2774.00	158.80	177.90	19.68	0.00
303300.00	2870.00	156.80	256.70	124.10	0.00
314300.00	2995.00	236.40	291.40	9.10	0.00
330500.00	3603.00	110.90	306.80	55.76	0.00
330500.00	3340.00	132.60	277.40	70.80	0.00
336700.00	4560.00	133.90	415.20	100.60	0.00
338100.00	3702.00	196.00	488.80	173.70	0.00
296700.00	3364.00	283.50	433.00	206.70	0.00
352000.00	5040.00	213.60	573.30	248.30	0.00
316400.00	4183.00	329.60	539.70	310.80	0.00
323800.00	3957.00	186.00	391.60	151.10	0.00
308400.00	4276.00	278.60	466.70	313.50	0.00
340600.00	3885.00	170.30	579.40	126.00	0.00
Average					
320113	**3146.05**	**203.24**	**282.37**	**154.21**	**0**

Table 7: Trace element results from Sa‘ad in ppm.

Ca	Sr	Zn	Mn	Pb	Al
171700.00	1178.00	285.70	73.27	0.00	680.80
171400.00	1525.00	547.60	126.80	0.00	136.10
188700.00	1073.00	361.30	78.53	0.00	649.10
188700.00	1628.00	774.90	178.30	0.00	1219.00
236200.00	1250.00	308.20	100.60	0.00	722.30
179200.00	1213.00	259.10	123.90	0.00	1139.00
180300.00	1413.00	494.70	141.40	0.00	1526.00
179300.00	1560.00	414.10	117.30	0.00	1174.00
187400.00	1476.00	489.20	127.50	0.00	1108.00
202800.00	1382.00	494.20	133.70	0.00	966.00
192000.00	1367.00	391.80	122.70	0.00	893.10
164400.00	1356.00	306.90	143.40	0.00	1728.00

Ca	Sr	Zn	Mn	Pb	Al
204200.00	1391.00	544.60	153.10	0.00	924.70
179900.00	1409.00	512.20	162.00	0.00	946.30
180900.00	1524.00	468.90	151.60	0.00	1264.00
207700.00	1592.00	465.40	161.20	0.00	959.50
198800.00	1213.00	379.80	162.50	0.00	1071.00
212600.00	1328.00	300.90	159.10	0.00	1640.00
177200.00	1689.00	461.60	221.70	0.00	1804.00
183700.00	1367.00	414.30	173.50	0.00	1852.00
Average					
189355	**1396.70**	**433.77**	**140.60**	**0.00**	**1120.14**

Table 8: Trace element results from Ya‘amun in ppm.

Ca	Sr	Zn	Mn	Pb	Al
176700.00	1229.00	193.90	178.50	0.00	2327.00
184200.00	962.80	125.40	151.70	0.00	4783.00
190500.00	986.50	157.50	186.70	0.00	1998.00
177200.00	1507.00	124.80	178.90	0.00	1725.00
208400.00	1442.00	158.50	137.20	0.00	1406.00
198900.00	1294.00	211.10	156.40	0.00	1840.00
173500.00	2541.00	148.20	227.10	0.00	2685.00
168300.00	1222.00	252.30	307.40	0.00	3016.00
158700.00	1356.00	190.80	173.60	0.00	1790.00
168200.00	1594.00	161.20	236.10	0.00	2120.00
209800.00	1459.00	279.30	199.80	0.00	2459.00
190600.00	1219.00	161.40	208.00	0.00	2274.00
177100.00	1430.00	152.30	229.50	0.00	2788.00
175600.00	1457.00	280.50	228.70	0.00	2479.00
177500.00	1409.00	157.60	230.80	0.00	2579.00
210900.00	1322.00	178.50	183.90	0.00	1867.00
181900.00	1621.00	212.10	328.60	0.00	3954.00

Ca	Sr	Zn	Mn	Pb	Al
186300.00	1100.00	138.50	272.50	0.00	2790.00
178900.00	1495.00	147.80	312.20	0.00	2907.00
193000.00	1391.00	159.00	239.10	0.00	3362.00
Average					
184310	**1401.87**	**179.53**	**218.33**	**0.00**	**2557.45**

Table 9: Trace element results from Juhfyiah in ppm.

Ca	Sr	Zn	Mn	Pb	Al
139400.00	1797.00	174.60	383.10	0.00	2773.00
189800.00	1817.00	245.70	210.20	0.00	2579.00
205300.00	2124.00	176.90	275.40	0.00	3115.00
205200.00	2004.00	111.40	249.70	0.00	3361.00
166000.00	2890.00	147.20	531.10	0.00	1.02
195200.00	1950.00	142.20	207.30	0.00	24466.00
178800.00	1734.00	179.90	203.10	0.00	2489.00
190000.00	2084.00	179.10	315.00	0.00	3016.00
201900.00	1822.00	179.50	229.30	0.00	2764.00
163200.00	2483.00	155.40	366.00	0.00	4052.00
187700.00	2353.00	167.50	272.70	0.00	3399.00
Average					
183863	**2096.18**	**169.03**	**294.80**	**0.00**	**4728.63**

Although the mineralogical composition of soil in the four sites is roughly the same (fig. 9), trace element exchange between soil and bone varies. That is probably attributed to the variations in types of burial (table 3), depth of the burial and water seepage as a factor of topography. The very high concentration of lead at the site of Bersenia might indicate the presence of lead coffins and/or lead artifacts inside the sampled tomb, a common burial practice during the Byzantine period (Abbadi 1960; Al-Shorman 2007). In addition, many leaded bronze rings and personal items were found in Byzantine burials (El-Najjar 2008).

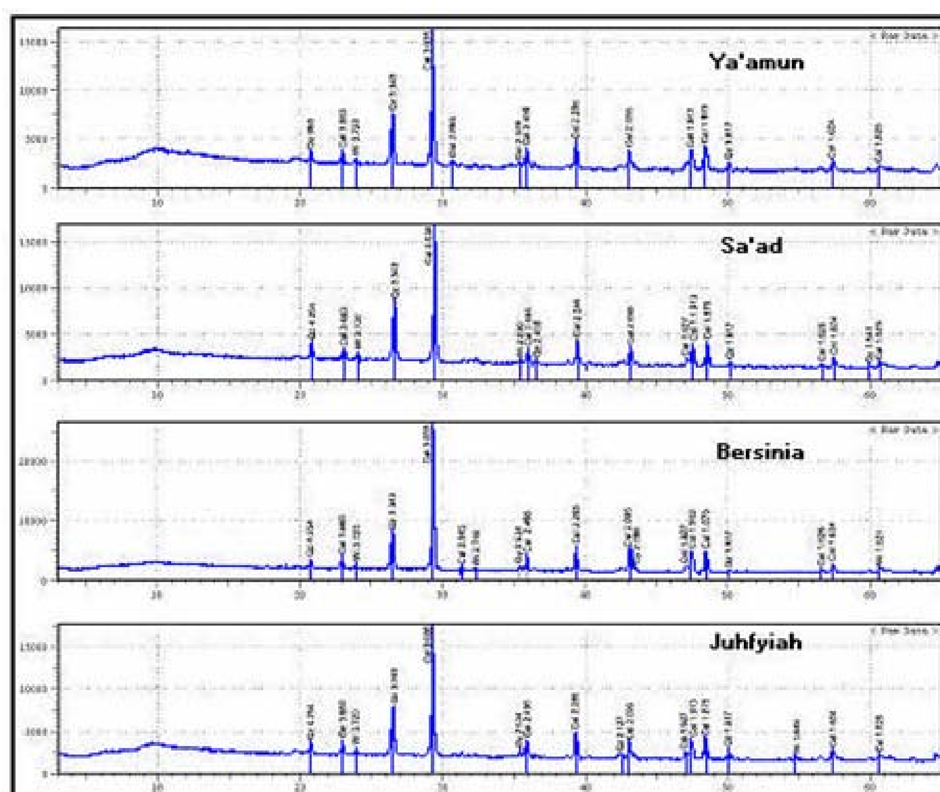


Fig. 3: X-ray diffraction of a soil samples from the four archaeological sites

Conclusions

Archaeological bones being sampled for reconstructions of paleodiet using trace element analysis should first be evaluated for diagenesis. The sampling procedure should take into account the burial type and topography to exclude the agents that may contribute to diagenesis. The burials sampled in this study are evidently not suitable for paleodietary reconstruction as the Ca/P ratioand the trace element concentrations are above the acknowledged standards. To overcome the impasse of diagenesis, teeth would be an appropriate alternative to extract trace elements and/or stable isotope analysis, either from bones or teeth. The interpretation of the chemistry results in archaeological bones, as is the case in this study, necessitates a broader knowledge of how the soil of the site was formed, where culture may change spatially and chronologically. In other words, the data is better understood within the frame of socioarchaeology.

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تلف البقايا العظمية من أربعة مواقع أثرية في شمال الأردن

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ملخـص

لقد تجاهلت دراسات كيمياء العظام الأثرية من المواقع الأردنية الدور المهم للتلف، حيث أدى ذلك إلى إحداث أخطاء في التفسير العلمي للعينات تحت الدراسة. ومن هنا جاءت هذه الدراسة لتقيم التلف في العينات العظمية الأثرية. وقد اشتملت الدراسة على ٨١ عينة عظمية إنسانية وحيوانية من أربعة مواقع بيزنطية في شمال الأردن. وتم تقييم التلف بناء على انحراف التركيبة الكيميائية للعظم عن عدد من المقاييس المعيارية لهذا الغرض، وذلك باستخدام أجهزة الحيود السينية والفلورة السينية والامتصاص الذري. ودلت الدراسة على وجود تلف واضح في العينات المدروسة مما يعيق مستقبلا عمل دراسات عن الغذاء القديم، وقد دلت أيضا على أن نوع المدفن هو أحد أهم العوامل التي تقلل من تلف البقايا العظمية، هذا بالإضافة إلى بيئة المدفن.

الكلمات الدالة: البقايا العظمية، المواقع الأثرية.

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